PATENT

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In re Application of:

Hiroshi NEMOTO et al.

Prior
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Prior
Examiner: Jonathan Crepeau

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For: LITHIUM SECONDARY BATTERY

PRELIMINARY AMENDMENT

Commissioner for Patents
Washington, D. C. 20231

Sir:

Prior to examination of the above application, please undertake the following changes:

IN THE SPECIFICATION:

Page 4, replace the third paragraph with:

According to the Battery Association of Japan, as a mechanical test (erroneous use test) of a lithium secondary battery safety estimation guideline, it is regulated that even if an abnormal discharge current abruptly flows by an internal short circuit of

electrodes which is caused by driving a nail (metal rod) in a surface (lamination surface) so that the nail pierces vertically electrode plates of a lithium secondary battery, which is fully charged in the charging capacity, the electrode plates being overlapped with each other on the surface, the battery does not burst, does not fire, and the safety can be secured (such a test will be hereinafter referred to as a nail piercing test).

Page 6, replace the first full paragraph with:

However, the relation between the battery capacitance and the opening area at the operation of the pressure release mechanism has not been clarified. That is, if the opening area at the operation of the pressure release mechanism is small, clogging occurs on the way to the pressure release of the internal pressure, so that the pressure release is not sufficiently carried out and there is a fear that such an accident as burst or firing of the battery would occur. On the other hand, if the opening area is large, although the fear of clogging can be removed, there is a fear that the constituents of the internal electrode body would jump out, or firing or combustion would occur when the internal electrode body jumps out in the state of the internal short circuit. However, conditions for preventing the occurrence have not been clear.

Page 10, second paragraph replace with:

As described above, according to the lithium secondary battery of the present invention, not to mention an internal short circuit, even in the case where a battery temperature is raised by overcharging due to an external short circuit or the like so that the internal pressure of the battery is increased, since the pressure release mechanism is disposed in a suitable shape for the battery capacitance and at a suitable place, the entire battery does not burst or explode, and superior safety is obtained. Moreover, even in the case where the pressure release mechanism functions also as a current path, since the pressure release mechanism is made of metal material members, the internal resistance of the battery is small and the battery is superior in charging and discharging characteristics.

Page 14, replace the second paragraph with:

As the electrolyte which is impregnated in the internal electrode body 90 and is filled in the battery case 63, it is preferable to use a nonaqueous organic electrolyte including a

single solvent or a solvent mixture of organic solvents such as ethylene carbonate, propylene carbonate, γ-butyrolactone, diethyl carbonate, tetrahydrofuran, and acetonitrile, and one or more kinds of $LiPF_6$, $LiClO_4$, $LiBF_4$, and lithium halide as an electrolyte dissolved in the solvent. Further, it is also possible to use a macromolecular solid electrolyte or the like formed by gelating and solidifying the thus formed electrolyte.

Replace the paragraph bridging pages 16 and 17 as follows:

When elastomer resin such as ethylene-propylene rubber is used for the thermal shrinkage tube 75, there are obtained such effects that the adhesion becomes excellent and the sealing becomes more complete. As the positive output terminal 76, the same material as the electrode material, such as aluminum or copper, is the most easily handled material. The shape of the positive output terminal is preferably such a flat ring shape that it is brought into uniform contact with the sealing portion of the battery case 63, and a terminal for connection to a load is protruded from a part thereof. One end of the conduction member 77 extending from the negative internal terminal 67 is electrically connected to the

negative output terminal 78 and is fixed to the insulating plate 73 by a screw 79 or the like.

Page 23, replace the first full paragraph with:

Though a pressure release mechanism is disposed in each of the end portions of the battery case 63 in the lithium secondary battery shown in Fig. 1, more than one pressure release mechanism may be disposed in each of the end portions of the battery case 63. For example, Fig. 10(a) is a plan view of conditions of disposing the metal foil 70 in Fig. 1 viewed from the direction of extension of the winding axis of the inner electrode body 90. In addition to such a structure, the safety equal to that obtained in the case employing the metal foil 70 can be secured as long as the area of opening portions is within the range satisfying $0.05 \leq S/C \leq 2$ even if more than one metal foil 42 is disposed as in Fig. 10(b).

Page 23, replace the third paragraph with:

By the way, when a pressure release mechanism in which the V-shaped groove 88 is used is formed on an end surface of a battery, it has a risk of breakage. In the case of not only Fig. 11(a) but

also Fig. 11(b), the pressure release mechanisms have a possibility of breakage by hitting the end surface provided with a V-shaped groove 88 of the battery against an obstacle. When an output terminal 44 is disposed inside the pressure release mechanism due to the V-shape groove 88, the V-shaped groove 88 is broken by external force applied to the output terminal 44 when batteries are connected with each other, thereby increasing a risk of breakage of the pressure release mechanism.

Page 25, first full paragraph replace with:

In the case of the lithium secondary battery using such a lamination-type internal electrode body, a pressure release mechanism is disposed at the side of the electrode plate, that is, at the side of the battery case corresponding to the side of the lamination surface of the internal electrode body in the outer circumferential direction. The reason for this configuration is the same as the case where in the lithium secondary battery using the wound-type internal electrode body, the pressure release mechanism is disposed at the end potion in the winding axis direction, that is, the end portion of the battery case corresponding to the side of the electrode plate.

Page 27, first full paragraph replace with:

An internal electrode body used for a lithium secondary battery satisfying this relation may be a wound type or a lamination type. When a wound-type internal electrode body is employed, the pressure release mechanism is disposed in at least one portion at one end of the battery case in the winding axis direction. When a lamination-type internal electrode body is employed, totally at least one pressure release mechanism is disposed on a side surface of the battery case perpendicular to a flat plate surface of the electrode plate or on each of at least two surfaces not facing to one another of the battery case.

Page 31, replace the third and fourth paragraphs with:

Hereinafter, description will be made to Example 1 and Example 2 with respect to a pressure release mechanism applied to a lithium secondary battery of the present invention, and Example 3 with respect to the relation between the battery capacity and the opening area of a pressure release mechanism.

First, Fig. 4 is an explanatory view of an apparatus 50 for carrying out an operation test (pressure withstand test) of a pressure release mechanism applied to the lithium secondary battery of the below Examples 1 and 2 of the present invention. As a sample 98, one having the structure of the pressure release mechanism shown in Fig. 2 is shown. However, as the sample 98, one in which the internal electrode body and electrode terminals were removed, was used.

Page 42, replace the first paragraph with:

From the test results of Table 3, in the both-end release-type battery, such a state was observed that in some batteries having an S/C value of less than 0.05, electrode materials and the like were clogged in the opening portion of the pressure release mechanism so that the battery case burst, or although the battery case did not burst seriously, a crack was produced at a portion other than the opening portion of the pressure release mechanism and gas spouted from this crack. On the other hand, when the S/C value is larger than 2, any problem did not occur in the operation of the pressure release mechanism, and since a nail was pierced in the internal electrode body, a part of the internal electrode body did not jump

out of the opening portion. However, in the case where an external short circuit occurs, there is a possibility that a part of the internal electrode body jumps out of the opening portion. When the S/C value is made large, the cylinder of the battery becomes large. Thus, when the battery is installed on an electric vehicle, a dead space for the arrangement becomes large, so that this is not preferable.

<u>IN THE CLAIMS</u>:

Cancel claim 1 without prejudice or disclaimer and substitute therefor the following:

40. (New) A lithium secondary battery comprising:

a battery case;

a negative electrode;

a lithium-containing positive electrode;

a separator, said negative electrode and said positive electrode being stacked or wound through said separator so that said negative electrode and said positive electrode are not in direct contact;

an organic electrolyte; and

a pressure release mechanism disposed on at least one end portion of said battery case, said pressure release mechanism comprising at least one sheet of a metal foil.

41. (New) The lithium secondary battery according to claim 40, wherein said at least one sheet of a metal foil is formed of a member selected from the group consisting of copper, nickel and aluminum.

42. (New) The lithium secondary battery according to claim 40, wherein said at least one sheet of a metal foil is an electrode plate for a negative electrode, and said pressure release mechanism further includes a metal sheet that is an electrode plate for a positive electrode.

43. (New) The lithium secondary battery according to claim 42, wherein said electrode plate for a negative electrode is copper or nickel, and said electrode plate for a positive electrode is aluminum, respectively.

44. (New) The lithium secondary battery according to claim 40, wherein an operational pressure of the pressure release mechanism is 2 to 10 kg/cm^2.

45. (New) The lithium secondary battery according to claim 40, wherein a difference in operational pressures of the installed pressure release mechanism is no more than 8 kg/cm^2.

46. (New) The lithium secondary battery according to claim 40, wherein the pressure release mechanism includes a groove portion disposed in a metal plate, the groove portion being burst so that an internal pressure of the battery is released to an outside pressure.

47. (New) The lithium secondary battery according to claim 40, wherein battery capacitance is not less than 5 Ah.

REMARKS

Applicants present for examination a further embodiment of the invention disclosed.

An early examination of claims 40 to 47 is earnestly solicited.

Respectfully submitted,

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MARKED UP SPEC

film are collapsed so that the movement of lithium ions is blocked and the battery reaction is suppressed.

Accordingly, also in a large capacity lithium secondary battery, it is conceivable that such various safety mechanisms as are installed in a small lithium secondary battery as described above must be indispensably installed.

However, since the resistivity of the above-mentioned PTC element made up of the conductive particles and polymer is about 1 Ω·cm at room temperature, the internal resistance of the battery becomes large to cause output loss, and the PTC element may cause the discharge characteristics to lower and the lifetime of the battery to shorten. Especially, in the case where such a PTC element is installed in a large capacity battery, the concentration of current in the inside of the PTC element is apt to occur due to the increase of an area of the PTC element, which causes heat generation, so that the installation to a large capacity battery is difficult. In addition, such a PTC element is generally expensive, and a large one in size is not manufactured, so that a current control element which is more inexpensive, is capable of dealing with a large battery, and has low resistance, is earnestly desired.

According to the Battery Association of Japan, as a mechanical test (erroneous use test) of a lithium secondary battery safety estimation guideline, it is regulated that even if an abnormal discharge current abruptly flows by an internal short circuit of electrodes which is caused by driving a nail (metal rod) in a surface (lamination surface) so that the nail pierces vertically electrode plates of a lithium secondary battery, which is fully charged in the charging capacity, the electrode plates being overlapped with each other on the surface, the battery does not burst, does not fire, and the safety can be secured (such a test will be hereinafter referred to as a nail piercing test).

releasing mechanism which operates without fail is related to the battery capacitance.

However, the relation between the battery capacitance and the opening area at the operation of the pressure release mechanism has not been clarified. That is, if the opening area at the operation of the pressure release mechanism is small, clogging occurs on the way to the pressure release of the internal pressure, so that the pressure release is not sufficiently carried out and there is a fear that such an accident as burst or firing of the battery would occur. On the other hand, if the opening area is large, although the fear of clogging can be removed, there is a fear that the constituents of the internal electrode body would jump out, or firing or combustion would occur when the internal electrode body jumps out in the state of the internal short circuit. However, conditions for preventing the occurrence have has not been clear.

Thus, there are often cases where the battery case itself becomes large since an unnecessarily large pressure release mechanism for battery capacity is disposed, or such restriction in configuration is imposed that although a battery of thin and long cylindrical shape is desired to be formed, a flat plate structure must be adopted while a battery capacity remains, since a large pressure release mechanism must be disposed.

Then the present inventors considered the structure, operational condition, and installation position of a pressure release mechanism for a large capacity lithium secondary battery with superior safety and low resistance, which is able to release a large pressure generated in the inside of the battery due to an electrode short circuit and the like to the atmospheric pressure, and as a result, the present invention has been achieved.

it is installed at the negative side. Moreover, it is preferable that an opening area of such a pressure release mechanism is 0.1 cm^2 or more.

Such structural condition of the lithium secondary battery of the present invention is preferably adopted for a battery having a battery capacitance of 5 Ah or more, and can be preferably used for an electric vehicle or a hybrid electric vehicle.

As described above, according to the lithium secondary battery of the present invention, not to mention an internal short circuit, even in the case where a battery temperature is raised by overcharging due to an external short circuit or the like so that the internal pressure of the battery is increased, since the pressure release mechanism is disposed in a suitable shape for the battery capacitance and at a suitable place, the entire of the battery does not burst or explode, and superior safety is obtained. Moreover, even in the case where the pressure release mechanism functions also as a current path, since the pressure release mechanism is made of metal material members, the internal resistance of the battery is small and the battery is superior in charging and discharging characteristics.

Brief Description of the Drawings

Fig. 1 is a sectional view showing an embodiment of a lithium secondary battery of the present invention.

Fig. 2 is a sectional view showing an embodiment of a pressure release mechanism adopted for the lithium secondary battery of the present invention.

Fig. 3 is a sectional view showing another embodiment of the pressure release mechanism adopted for the lithium secondary battery of the present invention.

and discharging reactions, so that the capacity of the battery is lowered. Thus, it is preferable to select a material in which the amount of the dead lithium is small.

As a material of the separator 62, it is preferable to use a three-layer structural material in which a polyethylene film having lithium ion permeability and including micropores is sandwiched between porous polypropylene films having lithium ion permeability. This serves also as a safety mechanism in which when the temperature of the internal electrode body 90 is raised, the polyethylene film is softened at about 130°C so that the micropores are collapsed to suppress the movement of lithium ions, that is, the battery reaction. When the polyethylene film is sandwiched between the polypropylene films having a softening temperature higher than the polyethylene film, it is possible to prevent the contact/welding between the separator film 62 and the positive and negative electrodes 60, 61.

As the electrolyte which is impregnated in the internal electrode body 90 and is filled in the battery case 63, it is preferable to use a nonaqueous organic electrolyte including a single solvent or a ~~mixture solvent~~ Solvent mixture of organic solvents, such as ethylene carbonate, propylene carbonate, γ-butyrolactone, ~~diethyle~~ diethyl carbonate, tetrahydrofuran, and acetonitrile, and one or more kinds of $LiPF_6$, $LiClO_4$, $LiBF_4$, and lithium halide as an electrolyte dissolved in the solvent. Further, it is also possible to use a macromolecular solid electrolyte or the like formed by gelating and solidifying the thus formed electrolyte.

Next, the structure of a negative side in the upper portion of the battery in Fig. 1 will be described. The negative electrode 61 is connected to leads 65 preferably at plural portions by resistance welding, ultrasonic welding, or the like. By carrying out the electricity collection from the plural portions in this way, as compared with the case where the electricity collection is carried out from only one portion, it

preferable to use copper, which is preferably used for the negative electrode material, for a material of the metal foil 70 in view of reactivity to the electrolyte, a nickel foil may be used.

An insulating plate 73 is disposed on the second seal ring 71, and the upper end of the battery case 63 is bent to be subjected to caulking, so that the negative internal terminal 67 and the like are fixed in the battery. The insulating plate 73 is provided with a communication hole 74 to release the internal pressure of the battery at the burst of the metal foil 70 at the pressure release hole 72 to the atmospheric pressure. A hard insulating resin such as Bakelite is preferably used for the material of the insulating plate 73. Instead of using the second seal ring 71, the insulating plate 73 may be modified to have the function of the second seal ring 71 so that the metal foil 70 is sandwiched between the first seal ring 69 and the insulating plate 73.

In order to insulate the negative internal terminal 67 from the battery case 63, a heat shrinkage tube 75 is inserted between the negative internal terminal 67 and the battery case 63. Moreover, as described later, since the battery case 63 functions as a current path at the positive side, a positive output terminal 76 is inserted and fixed between the battery case 63 and the heat shrinkage tube 75 at the bent position of the upper end of the battery case 63. A conductive member 77 for connecting the negative internal terminal 67 to the outside is disposed at a part of the outer circumference of the insulting plate 73, and is assembled and fixed so as not to come in contact with the positive output terminal 76, whereby the negative side of the battery case 63 is sealed (made airtight).

When elastomer resin such as ethylene-propylene rubber is used for the thermal shrinkage tube 75, there are obtained such effects that the adhesion becomes excellent and the sealing becomes more complete. As the positive output terminal

76, the same material as the electrode material, such as aluminum or copper, is the most easily handled material. The shape of the positive output terminal is preferably such a flat ring shape that it is brought into uniform contact with the sealing portion of the battery case 63, and a terminal for connection to a load is protruded from a part thereof. One end of the conduction member 77 extending from the negative internal terminal 67 is electrically connected to the negative output terminal 78 and is fixed to the insulating plate 73 by a screw 79 or the like.

Next, the structure of a positive side in the lower portion in Fig. 1 will be described. The positive electrode 60 and leads 80 are connected to each other at plural portions by resistance welding, ultrasonic welding or the like. The reason why the electricity collection is carried out from the plural portions in this way is the same as the electricity collection at the negative electrode 61. As a material for the leads 80, aluminum identical to the material of the positive electrode 60 is preferably used.

These leads 80 attached to the positive electrode 60 are connected to an aluminum rivet 81 and electricity collection is made. The rivet 81 is integrated with a positive internal terminal plate 82 of aluminum by welding.

The positive internal terminal plate 82 is positioned by a caulking portion 84 provided on the battery case 63 through an insulating heat shrinkage tube 83 so as to prevent the positive internal terminal plate from coming in direct contact with the inner surface of the battery case 63. A part of the positive internal terminal plate and an aluminum battery cap 85 as a bottom of the battery forms contact A through welding. Further, the positive internal terminal plate 82 is provided with a communication hole 86, so that the internal pressure of the battery is sensed by the battery cap 85.

battery case must be large is added, there may occur an undesirable case for designing to save a mounting space of the battery.

Though a pressure release mechanism is disposed in each of the end portions of the battery case 63 in the lithium secondary battery shown in Fig. 1, more than one pressure release mechanism~~s~~ may be disposed in each of the end portions of the battery case 63. For example, Fig. 10(a) is a plan view of conditions of disposing the metal foil 70 in Fig. 1 viewed from the direction of extension of the winding axis of the inner electrode body 90. In addition to such a structure, the safety equal to that obtained in the case employing the metal foil 70 can be secured as long as the area of opening portions is within the range satisfying 0.05 ≤ S/C ≤ 2 even if more than one metal foil~~s~~ 42 ~~are~~ is disposed as in Fig. 10(b).

A plurality of pressure release mechanisms are provided in the same manner as in the case employing the V-shaped groove 88. In this case, a required pressure release area may be secured by arranging pressure release mechanism due to a plurality of V-shaped grooves 43 as shown in Fig. 11(b) instead of forming a V-shaped groove 88 in a shape of a circle as shown in Fig. 11(a).

By the way, when a pressure release mechanism in which the V-shaped groove 88 is used is formed on an end surface of a battery, it has a risk of breakage. In the case of not only Fig. 11(a) but also Fig. 11(b), the pressure release mechanisms ~~has~~ have a possibility of breakage by hitting the end surface provided with a V-shaped groove 88 of the battery against an obstacle. When an output terminal 44 is disposed inside the pressure release mechanism due to the V-shape groove 88, the V-shaped groove 88 is broken by external force applied to the output terminal 44 when batteries are connected with each other, thereby increasing a risk of breakage of the pressure release mechanism.

plates 8 and 9, and the number of laminated plates. However, since a lead line 6 is required for the respective electrode plates 8 and 9, the inside of the battery becomes complicated so that the lamination- type is inferior to the wound-type in view of the assembling operation of the battery. Incidentally, when such lamination-type internal electrode body 7 is used, instead of the separator 10, it is also possible to use a solid electrolyte of an organic or inorganic material having functions of both the separator 10 and the electrolyte.

In the case of the lithium secondary battery using such a lamination-type internal electrode body, a pressure release mechanism is disposed at the side of the electrode plate, that is, at the side of the battery case corresponding to the side of the lamination surface of the internal electrode body in the outer circumferential direction. The reason of this configuration is the same as the case where in the lithium secondary battery using the wound-type internal electrode body, the pressure release mechanism is disposed at the end portion in the winding axis direction, that is, the end portion of the battery case corresponding to the side of the electrode plate.

At this time, in general, if the capacity of the battery is the same, it is preferable to make the volume small, and also it is preferable to design the structure so that serial and parallel connection of batteries is easy. Thus, it is preferable to dispose a pressure release mechanism and an output terminal respectively on the side portions opposite to each other with respect to the center of a flat surface of an electrode plate. For example, if the internal electrode body is shaped into a rectangular parallelepiped, such an example may be shown that both the output terminal and the pressure release mechanism are disposed on each of a pair of opposite sides of a rectangular parallelepiped battery case, and a superfluous space

secure the safety. In this case, design is made so that when the battery capacity is C (Ah), and the total area of opening portion where the pressure release mechanism operates is S (cm^2), the relation of $0.5 \leq S/C \leq 2$ is established. Although the structure of an internal electrode body in this case may be of the wound-type or lamination-type, the position of the disposed pressure release mechanism is the end or side of the battery case corresponding to the side of the electrode plate.

An internal electrode body used for a lithium secondary battery satisfying this relation may be a wound type or a lamination type. When a wound-type internal electrode body is employed, the pressure release mechanism is disposed in at least one portion at one end of the battery case in the winding axis direction. When a lamination-type internal electrode body is employed, totally at least one pressure release mechanism is disposed on a side surface of the battery case perpendicular to a flat plate surface of the electrode plate or on each of at least two surfaces not facing to one another of the battery case.

In this case, the lower value of S/C is 0.5 which is larger than the case where the pressure release mechanisms are disposed at two portions, and like this, when the opening area of the pressure release mechanism is made large with respect to the battery capacity, release of the internal pressure becomes easy. As a result, it is possible to decrease the load of pressure to the end where the pressure release mechanism is not disposed, and it becomes possible to prevent the burst or firing of the battery. This condition is also a condition that the opening area of the pressure release mechanism restricts the shape of the battery. That is, according to the battery capacity, according to the value of S/C, a minimum area of an end surface or a side of a battery case which is necessary to dispose the pressure release mechanism having a certain opening area, is determined, and as a result, the length

Although the structural conditions and pressure release mechanism of the lithium secondary battery of the present invention have been described, it is needless to say that the same pressure release mechanism may be adopted for both electrode portions of one battery, or a pressure release mechanism of different structure may be arbitrarily selected. Moreover, an output terminal for each of positive and negative electrodes may be disposed at each end of the battery, while positive and negative electrodes are concentrically disposed at one end of the battery.

The structural condition of the thus fabricated lithium secondary battery of the present invention is preferably applied to a large capacity battery with a battery capacity of 5 Ah or more, and such a large capacity battery is preferably used as a battery for an EV and an HEV. However, it is needless to say that the present invention can also be used for the structure of a small battery.

Examples

Hereinafter, description will be made to example 1 and example 2 with respect to a pressure release mechanism applied to a lithium secondary battery of the present invention, and example 3 with respect to the relation between the battery capacity and the opening area of a pressure release mechanism.

First, Fig. 4 is an explanatory view of an apparatus 50 for carrying out an operation test (pressure withstand test) of a pressure release mechanism applied to the lithium secondary battery of the below examples 1 and 2 of the present invention. As a sample 98, one having the structure of the pressure release mechanism shown in Fig. 2 is shown. However, as the sample 98, one in which the internal electrode body and electrode terminals were removed, was used.

From the test results of Table 3, in the both-end release-type battery, such a state was observed that in some batteries having an S/C value of less than 0.05, electrode materials and the like were clogged in the opening portion of the pressure release mechanism so that the battery case burst, or although the battery case did not burst seriously, a crack was produced at a portion other than the opening portion of the pressure release mechanism and gas spouted from this crack. On the other hand, when the S/C value is larger than 2, any problem did not occur in the operation of the pressure release mechanism, and since a nail was pierced in the internal electrode body, a part of the internal electrode body did not jump out of the opening portion. However, in the case where an external short circuit occurs, there is a possibility that a part of the internal electrode body jumps out of the opening portion. When the S/C value is made large, the cylinder of the battery becomes large. Thus, when the battery is installed on an electric vehicle, a dead space for the arrangement becomes large, so that this is not preferable.

On the other hand, in the single-end pressure-release type battery, in the case where the value of S/C was less than 0.5, it was observed that the battery case burst at a portion of the end where the pressure release mechanism was disposed, other than the pressure release mechanism, or at the end where the pressure release mechanism was not disposed, and vapor, which was presumed to be a product formed by evaporation of the electrolyte, and the electrolyte spouted. In the case where the value of S/C is larger than 2, similarly to the case of the both-end release-type battery, although there arose no problem on the operation of the pressure release mechanism, it was observed that the internal electrode body moved slightly to the side of the opening portion. This suggests a possibility that a part of the internal electrode body jumps out of the opening portion of the pressure release mechanism.